

Dephasing of Mollow Triplet Sideband Emission of a Resonantly Driven Quantum Dot in a Microcavity

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Abstract

Detailed properties of resonance fluorescence from a single quantum dot in a micropillar cavity are investigated, with particular focus on emission coherence in dependence on optical driving field power and detuning. Power-dependent series over a wide range could trace characteristic Mollow triplet spectra with large Rabi splittings of $|\Omega| \leq 15$ GHz. In particular, the effect of dephasing in terms of systematic spectral broadening $\propto \Omega^2$ of the Mollow sidebands is observed as a strong fingerprint of *excitation-induced dephasing*. Our results are in excellent agreement with predictions of a recently presented model on phonon-dressed QD Mollow triplet emission in the cavity-QED regime.

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Exploiting the quantum properties of the light which is emitted from semiconductor quantum dots (QDs) has the potential of enabling various new applications in the field of photonics and quantum information technology [1]. Many of these applications require single-[2, 3] and entangled-photon [4–7] light sources. The implementation of single-photon based quantum logic algorithms critically relies on photon indistinguishability [8] which is directly related to the coherence properties of the emitted light. The coherence time T_2 can be defined via the excited state’s dephasing rate $(T_2)^{-1} = (2T_1)^{-1} + (T_2^*)^{-1}$, with T_1 as the radiative emitter lifetime and T_2^* as the pure dephasing time. True resonant s-shell excitation appears beneficial to avoid pure dephasing, and is therefore highly anticipated to approach the ideal Fourier transform limit [9, 10] given by $T_2/(2T_1) = 1$. The observation of the Mollow triplet is the characteristic hallmark of resonance fluorescence from a strongly driven and dressed two-level system. Resonance fluorescence emission from a single QD has been first reported by Muller et al. [11]. Recently, they also directly observed Mollow triplets [12] with Rabi splittings up to $\Omega \approx 4.4$ GHz ($18 \mu\text{eV}$), where a distinct pump-power independent dephasing rate of $\Gamma_2^* = (T_2^*)^{-1} \approx 3.1$ GHz has been found [13]. In another remarkable work by Vamivakas *et al.* [14] the resonance fluorescence and spin-resolved characteristics of a trionic QD state have been investigated. The Mollow triplet sideband emission showed a high degree of coherence with $\Gamma_2^* \approx 18$ MHz. Important to note, all these investigations have been performed under moderate pumping levels ($\Omega < 6$ GHz) and investigations under stronger s-shell excitation have not been reported so far.

In this letter, we report on detailed investigations of resonance fluorescence emission from single QD neutral excitonic X^0 recombination in a high- Q micropillar cavity. The influence of (a) *resonant pump power* and (b) *frequency detuning of the driving laser* ($-3 \text{ GHz} < \Delta < 4 \text{ GHz}$) on the characteristic Mollow sideband emission is investigated, with particular focus on the coherence properties of the emission.

The sample structure under study is grown by molecular beam epitaxy on GaAs substrate. The initial planar cavity consist of 30 (26) AlAs/GaAs distributed Bragg reflector period pairs as the bottom (top) mirrors, respectively. Spacer of 2×130 nm GaAs form a λ -cavity around a single centered layer of (In,Ga)As QDs with a spatial density of $\sim 6 \cdot 10^9 \text{ cm}^{-2}$. Ordered fields of different diameter ($1.5 - 4 \mu\text{m}$) high-quality micropillars have been finally processed by combined electron-beam lithography and plasma-induced reactive ion etching [15].

Our investigations are performed on a confocal low-temperature ($T \geq 4$ K) micro-photoluminescence (μ -PL) setup [10], using a special orthogonal symmetry between lateral optical laser excitation of individual micropillar structures close to the cleaved [110] sample edge and vertical QD emission detection along the pillar symmetry axis. A narrow-band (FWHM ≈ 500 kHz) continuous-wave (cw) Ti:Sapphire ring laser or by a mode-locked Ti:Sa pulse laser (~ 2 ps pulses at $f_{rep} = 76.2$ MHz) for time-resolved μ -PL (TCPC: time-correlated photon counting) are used for excitation. In addition to μ -PL detection by a spectrometer/CCD system ($\Delta E_{res} \sim 35 \mu\text{eV}$ resolution), a scanning Fabry-Pérot interferometer [10] (Finesse $F \sim 150$; FSR = 15 GHz $\sim 62.035 \mu\text{eV}$ free spectral range) provides high-resolution PL (HRPL) with $\Delta E_{res}^{\text{HRPL}} < 1 \mu\text{eV}$.

The presented measurements have been performed on a $1.75 \mu\text{m}$ diameter micropillar. As shown in Fig. 1(a), this microcavity reveals single-QD neutral exciton X^0 s-shell emission at ~ 1.3571 eV close to the FM (~ 1.3568 eV) at $T = 10$ K under selective *quasi-resonant* QD p-shell excitation ~ 22 meV above the ground state. At this QD-FM detuning $\Delta E = +280 \mu\text{eV}$, the spontaneous radiative X^0 decay time has been measured as $\tau_{dec} = 820 \pm 40$ ps by TCPC under pulsed p-shell excitation (see inset of Fig. 1(a)). Due to very fast carrier relaxation between the p- and s-shell in those QDs (typ. on a few ps-scale), the pure radiative lifetime T_1 is expected to obey $T_1 \approx \tau_{dec}$. From the ratio of FM emission energy and linewidth δE (FWHM), we derive a high quality factor of $Q \approx E/\delta E = 13500 \pm 500$ for this micropillar. Worth to note, the prominent effect of fundamental mode emission despite its distinct spectral *detuning* from the dominantly 'feeding' single QD is due to *non-resonant QD-mode coupling* in such solid-state emitter-cavity systems [16–19].

As was first theoretically described by B. R. Mollow [12], a strong and resonant driving light field (with detuning $\Delta = 0$) 'dresses' the electronic states of a two-level system into a four-level system (Fig. 2(a)). Optical emission of such a system is described by characteristic multi-Lorentzian *Mollow triplet* spectra, composed of a central peak at the bare emitter frequency ν_0 , symmetrically decorated by two satellites at $\nu_0 \pm \Omega$ (see Fig. 2(b)). $\Omega = \mu E_0/\hbar$ represents the bare Rabi frequency (in Hz) with transition dipole moment μ and the local field strength E_0 at the emitter. For $\Delta = 0$ and sufficient excitation, the energetic sideband splitting with respect to ν_0 is given by $\sim |\Omega|$, i.e., obeys a proportionality to the square root of excitation power $P_0^{1/2}$.

Results of systematic HRPL investigations on single QD exciton s-shell emission under

strictly resonant ($\Delta = 0$), power-dependent cw excitation are depicted in Fig. 2(b). For enhanced clarity, the spectra are vertically shifted. Spectral detuning of emission relative to the bare X^0 transition is denoted as δ in units of frequency (GHz) and energy (μeV). As a consequence of the Fabry-Pérot technique used in HRPL, all spectral emission features appear periodically with an offset equal to the interferometer FSR. With increasing power, the evolution of two symmetrically spaced sidebands can be clearly observed around the central line which is composed of QD resonance fluorescence and residual scattered laser light. By variation of the pump power over more than two orders of magnitude, a systematic increase of Rabi sideband splittings up to $\Omega \approx \pm 15$ GHz ($\pm 62\mu\text{eV}$) is traced, limited only by the FSR of our interferometer, as here the sidebands start to overlap with adjacent FPI transmission orders. Extracted side peak splittings $|\Omega|$ from Fig. 2(b) are plotted in Fig. 2(c) as a function of the square root of the driving laser power ($\sim P_0^{1/2}$). The theoretically expected proportionality is clearly confirmed, as demonstrated by the applied linear fit (solid line). We like to emphasize that in our studies the investigated excitation power range and consequently the regime to observe Rabi splitting values of $|\Omega|$ is significantly larger than in previously reported investigations [13, 14] using direct detection of 'dressed' state fluorescence from single QDs.

From a more detailed inspection of the Mollow triplet series in Fig. 2(b), we particularly observe also a systematic and distinct spectral broadening of the Rabi sidebands with increasing pump strength. For preliminary analysis, the spectral line widths $\Delta\nu$ and their gradual broadening have been deduced from Lorentzian least-square fits to the HRPL data (not shown). Figure 2(d) (black trace) depicts corresponding FWHM values (GHz) of the Rabi sidebands, which reveal an overall line width increase by factor ~ 1.8 over the observed power range. In particular, a clear linear dependence $\Delta\nu \propto \Omega^2$ on the squared Rabi frequency is traced, which represents a strong indication of *excitation-induced dephasing* (EID) as an important additional effect accompanying the 'dressed' character of resonant QD emission. As is shown below, our experimental findings are in full quantitative consistence with predictions of a recently presented model on phonon-dressed QD Mollow triplet emission in the cavity-QED regime [20].

Prior to detailed data analysis, we note here that other studies on *pulsed resonant* single QD excitation have interpreted the effect of EID in terms of (time-domain) Rabi rotation damping to originate from coherent energy exchange between the emitter and a resonant LA-

phonon bath in the barrier matrix [21, 22]. In addition to pulsed broadband emission, their model addressed InGaAs QDs in $n-i$ -Schottky diode structures *without* a surrounding micro cavity – in clear contrast to our experimental conditions. Alternatively, carrier scattering between a resonantly *pulsed* single QD in a photodiode structure (without cavity coupling) and off-resonant wetting layer and multi-exciton states were theoretically discussed as the origin of EID with respect to decoherence in Rabi oscillations [23]. Moreover, for resonance fluorescence under pure *continuous wave* (cw) conditions, photon statistics and emission dephasing via phonon-bath coupling of a single bare QD without cavity coupling have also been analyzed theoretically [24]. In qualitative consistence of all previous studies [21–24], EID rates proportional to the squared Rabi frequency Ω^2 were concluded. Nevertheless, the particular regime of cavity-QED providing distinct emitter-mode coupling between a single ‘dressed’ QD and a high-Q 3D micro cavity has not been addressed by either previous study, thus hindering a direct numerical interpretation of our data in Fig. 2. In contrast, we will compare our results to a new recently presented theory by Roy and Hughes [20] on cw-excited single QD resonance fluorescence in a high-Q cavity, which explicitly considers the combined effects of electron-acoustic phonon bath coupling and electron-photon coupling to model a full *cavity-QED system* on the basis of polaronic dynamics.

To quantitatively interpret the observed spectral sideband broadening (Fig. 2(b)), our Mollow triplet spectra were modeled on the theoretical basis of Ref. 13. Although the authors report no EID effect from their experiments, the influence of pure dephasing was already accounted for by independent rates of radiative decay $(2T_1)^{-1}$ and decoherence $\Gamma = T_2^{-1}$ (Ref. 13, Eqn. 3). In our analysis, the total dephasing rate was substituted as $\Gamma = (2T_1)^{-1} + K \cdot \Omega^2$ to compute the side peak spectra in dependence on *radiative dephasing* and power-dependent *pure dephasing* (EID) with emission broadening $\Delta\nu \propto \Omega^2$. For various fixed values of K (GHz^{-1}), the whole set of Mollow triplet side peak spectra was repeatedly calculated [25]. High consistence over the full power series could be achieved for a dephasing parameter $K = 0.005 \pm 0.001 \text{ GHz}^{-1}$ (at $T = 10 \text{ K}$), well reproducing all side peak spectra (Fig. 2(b), bold red lines) and their overall FWHM broadening, i.e. the according dephasing rate Γ extracted in Fig. 2(d). Worth emphasizing, apart from radiative dephasing no extra constant pure dephasing needed to be included, which anticipates close to ideal Fourier transform-limited resonance fluorescence $T_2/(2T_1) \approx 1$ in the limit $\Omega^2 \rightarrow 0$, in full agreement with our previous studies on single-QD resonance emission [10].

To numerically compare the observed Mollow side peak broadening with respect to predictions of the QED model and the coupling regime considered in Ref. 20, we deduced the emitter-cavity coupling strength g for the studied QD-cavity system from $g \approx \sqrt{\frac{F_P \cdot \kappa_{cav}}{4\tau_X}}$. For a measured Purcell emission enhancement factor of $F_P \approx 13$, a FM cavity photon loss rate of $\kappa = \frac{2\pi c_0}{Q \cdot \lambda_X} \approx 104 \mu\text{eV}$ (i.e. ~ 25.2 GHz) and the measured excitonic emission life time of $\tau_X = 820 \pm 40$ ps at $\Delta E = +280 \mu\text{eV}$ QD-FM detuning (Fig. 1(a)), we obtain a value of $g \approx 16.2 \pm 0.5 \mu\text{eV}$ (or 3.9 ± 0.1 GHz), conform with the regime described by Roy and Hughes [20]. Their calculations, explicitly considering pure cw-resonant s-shell excitation under zero laser detuning ($\delta = 0$), clearly predict the effect of excitation-induced dephasing (EID) due to combined emitter phonon-bath coupling and cavity-photon bath interaction especially in those cavity QED systems. In particular, distinct spectral Mollow triplet side band broadening $\Delta\nu \propto \Omega^2$ is expected for small and moderate emitter-cavity couplings in the range $15 \mu\text{eV} \leq g \leq 50 \mu\text{eV}$. Considering parameters very similar to those valid for our experiments, Mollow side peak bandwidth enhancement with $K \approx 0.005 - 0.007 \text{ GHz}^{-1}$ (equivalent to $K \approx 0.001 - 0.002 \mu\text{eV}^{-1}$) is indeed anticipated, in high quantitative conformity with our experimental results discussed above (Fig. 2(d)).

Furthermore, as a direct consequence of phonon-mediated non-resonant cavity feeding from the detuned QD into the detuned FM ($\Delta E = +280 \mu\text{eV}$ here), an asymmetry between the Mollow triplet side band emission intensities is expected [20] due to their slightly different spectral detuning to the micro cavity mode. Clear indications of this effect could be consistently traced from the detailed theoretical fit of our HRPL series in Fig. 2(a) (bold lines), reflecting $\sim 15\%$ higher intensity from the low frequency Mollow triplet side bands at $\nu_0 - \Omega$ (i.e. spectrally closer to the FM) with respect to their counterparts at $\nu_0 + \Omega$.

The second part of our studies focused on single-QD Mollow triplet spectra in explicit dependence on laser frequency detuning. According to theory [14], non-zero spectral detuning Δ between the driving field and the bare emitter resonance ν_0 characteristically modifies the 'dressed' emission. Besides the center transition at $\nu_0 + \Delta$, the two sideband frequencies become $\nu_0 + \Delta \pm \Omega'$, where Ω' denotes the *generalized Rabi frequency* $\Omega' = \sqrt{\Omega^2 + \Delta^2}$ with Ω as the (zero detuning) 'bare' Rabi frequency at a given excitation power.

Figure 3(a) depicts HRPL spectra at a fixed laser power of $P_0 \approx 16 \mu\text{W}$, taken under systematic detuning of the excitation frequency from the QD s-shell resonance over a large range of $-3 \text{ GHz} \leq \Delta \leq +4 \text{ GHz}$ (indicated by arrow markers). The center peak composed

of QD resonance fluorescence and intense laser stray light has been removed from the spectra for clarity. Under variation of Δ , a gradual shift of the whole Mollow triplet signature and a significant increase of sideband splitting with increasing detuning are clearly observable. Figure 3(b) extracts the peak positions of each emission component in Fig. 3(a) as a function of spectral laser detuning Δ . As a reference, the horizontal dashed line denotes the symmetry point $\Omega' = \Omega$ of zero-detuning from the QD s-shell. Explicit fits of the sideband positions according to $\nu(\Delta) = \nu_0 + \Delta \pm \sqrt{\Omega^2 + \Delta^2}$ (Fig. 3(b), bold red lines) reveal high consistence with theory. For a more convenient analysis of the detuning series, values of the *full sideband splittings* as a function of Δ are plotted in Fig. 3(c), revealing a minimum for $\Delta \rightarrow 0$. From a least-square fit $\propto 2\sqrt{\Omega^2 + \Delta^2}$ to the data (bold line) at this fixed power level, we derive a bare Rabi frequency of $\Omega = 3.99 \pm 0.06$ GHz. Taking into account this value of Ω together with idealized beam geometry and spatial QD-field overlap for an estimation of the local field strength $|E_{loc}|$, we deduce the magnitude of the electric dipole moment of this particular QD as $\mu_{el} = \hbar\Omega/|E_{loc}| = 18 \pm 2$ Debye. This value is only somewhat smaller than recently reported values for neutral X^0 states in similar types of InAs/GaAs QDs [14].

Another interesting phenomenon is traced from a detailed evaluation of the sideband FWHM in the HRPL series of Fig. 3(a), extracted in Fig. 3(d) (color-coded for the red (blue) detuned Mollow triplet side bands). Under increasing laser detuning from the QD s-shell resonance but fixed excitation power ($P_0 \approx 16 \mu\text{W}$), we observe a distinct spectral *narrowing* of either side band with increasing Δ . Worth noting, this behaviour appears unexpected in terms of the cavity QED-based EID model [20], as 'dressed' emission with increased side band splittings equivalent to the generalized Rabi frequency $\sqrt{\Omega^2 + \Delta^2}$ should reflect also in increased dephasing [26], i.e. line broadening $\propto \Omega'$ – in contrast to our experiment. On the other hand, first indications of aberrations from the above denoted theoretically expected emission detuning dependence are found by detailed inspection of the extracted total side band splitting with Δ in Fig. 3(c). With increasing detuning from resonance we observe an increasing deviation between the measured splittings and the theoretical model, which might tentatively be interpreted to result from a systematically reducing bare Rabi frequency independent of the increasing value of Δ . Nevertheless, a fundamental interpretation of this effect has to be left for further ongoing in-depth experimental and theoretical analysis.

In conclusion, resonance fluorescence emission properties of a single InAs/GaAs QD in a high-quality micropillar cavity have been investigated in detail with particular focus on

excitation power and/or *excitation detuning* relative to the s-shell ground state. Wide-range excitation power series could trace characteristic Mollow triplet spectra with large Rabi splittings of $\Omega \approx \pm 15$ GHz, accompanied by the effect of systematic spectral sideband broadening $\propto \Omega^2$ as a strong indication of *excitation-induced dephasing* (EID).

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Figures:

Fig. 1

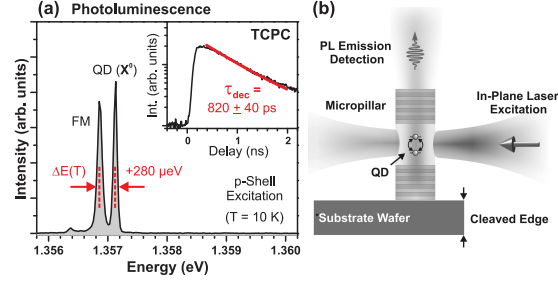


Fig. 2

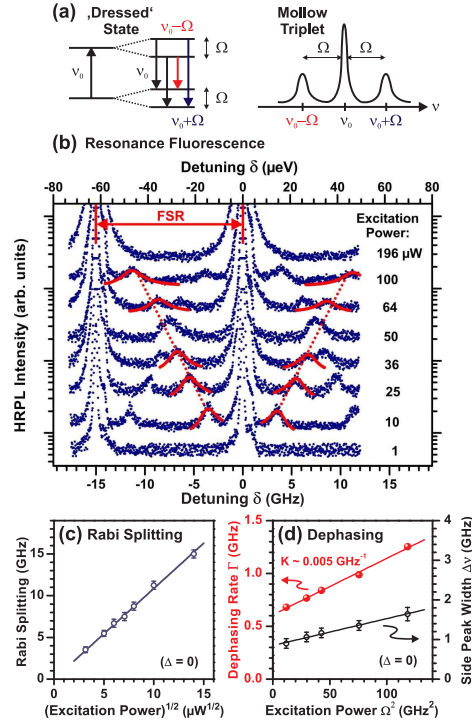


Fig. 3

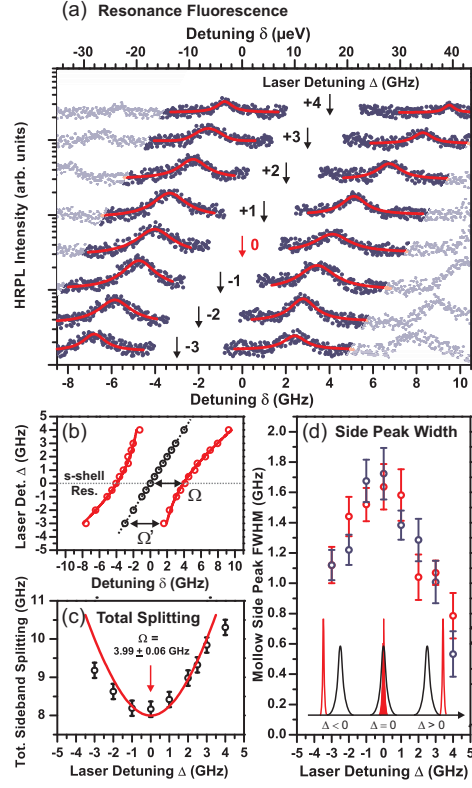


Figure Captions:

Fig. 1 (color online) (a) Single QD neutral exciton (X^0) emission spectrally close to the fundamental mode (FM) of a $1.75\ \mu\text{m}$ micropillar cavity, observed at quasi-resonant p-shell excitation ($P_0 \approx 50\ \mu\text{W}$; $\Delta E(T)$: temperature-dependent QD-mode detuning). Inset: Time-resolved X^0 emission signal. (b) Schematic geometry of orthogonal excitation and detection.

Fig. 2 (color online) (a) Energy scheme (left) and characteristic Mollow triplet spectra (right) of a 'dressed' two-level system. (b) Power-dependent high-resolution (HRPL) spectra of X^0 emission at pure resonant ($\Delta = 0$) continuous-wave excitation. (c) Mollow sideband splitting extracted from plot (b), revealing the expected linear dependence on the square root of laser power $|\Omega| \sim P_0^{1/2}$ (GHz). (d) Power-dependent Mollow sideband FWHM $\Delta\nu$ and total dephasing rate $\Gamma \propto \Omega^2$ from theoretical analysis of data in (a), indicative of excitation-induced dephasing (EID).

Fig. 3 (color online) (a) HRPL of QD resonance fluorescence under excitation detuning Δ (see arrows) relative to the QD s-shell, taken at a fixed power of $P_0 = 16\ \mu\text{W}$. Red traces: Lorentzian FWHM fits. (b) Spectral evolution of Mollow sidebands (red) with laser detuning Δ (black center trace), derived from (a). (c) Total sideband splitting in plot (b), together with a theory fit to evaluate the bare Rabi frequency Ω . (d) Sideband FWHM vs. laser detuning Δ from plot (a), revealing the phenomenon of FWHM reduction with increasing Δ . Please note a reduced QD-FM detuning $\Delta E = +230\ \mu\text{eV}$, yielding $T_1 \sim 410 \pm 20\ \text{ps}$ and correspondingly larger FWHM by factor ~ 1.6 for $\Delta = 0$ conditions with respect to Fig. 2(d).